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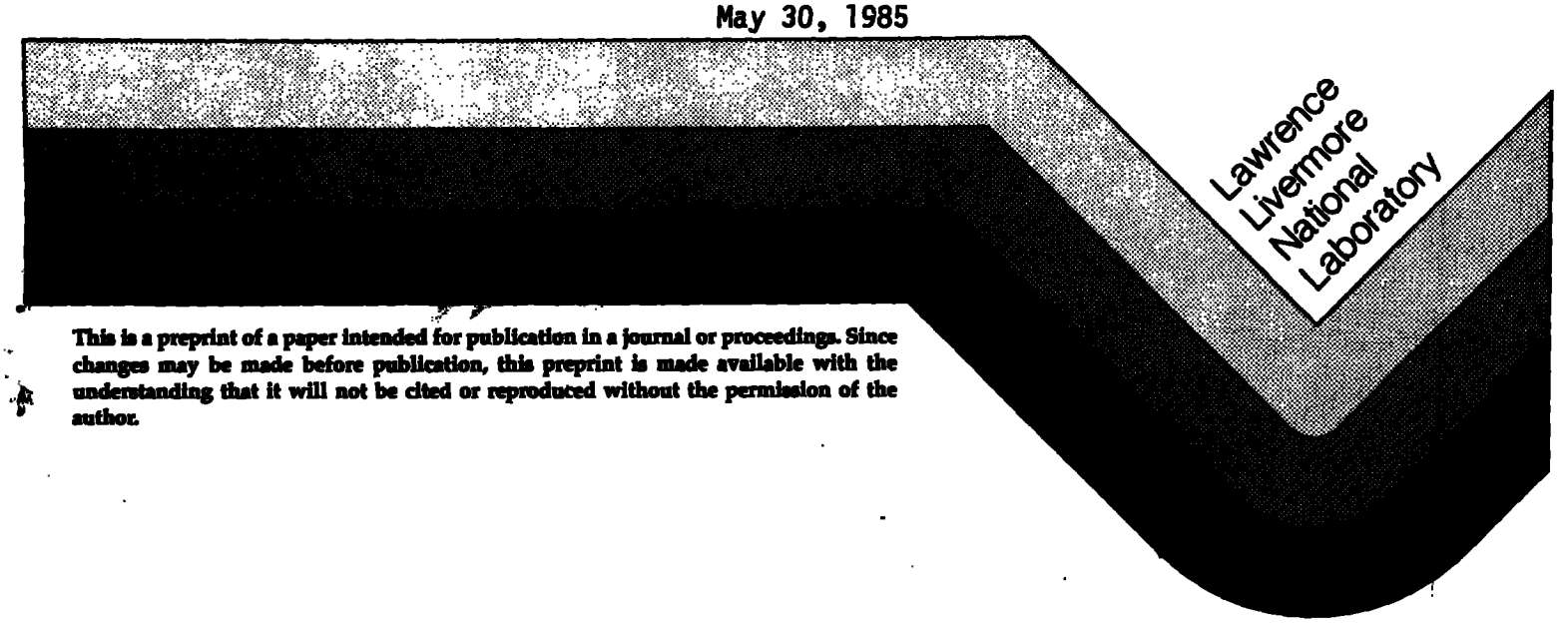

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Pulse Duration Dependence of 1064-nm Laser Damage Thresholds of Porous Silica Antireflection Coatings on Fused Silica Substrates*

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Abstract

We used 1064-nm pulses with durations of 1, 5, 9 ns to measure laser-damage thresholds of 10 porous silica antireflection coatings deposited from both methanol and ethanol solutions containing silica particles with diameters of 10-20 nm. The median thresholds measured at the three pulse durations, 10.8 J/cm^2, 26.5 J/cm^2, and 38 J/cm^2, scaled as pulse duration to the 0.56 power.

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1. Introduction

Porous silica coatings were developed at Livermore in response to the need for damage resistant antireflection films for use at ultraviolet wavelengths, primarily 248 nm and 351 nm. Because these coatings performed well in the ultraviolet, and because techniques had been developed for applying such films to optical elements with diameters up to 1 meter, it was interesting to evaluate their potential for use at 1064 nm.

2. Fabrication of Test Coatings

The coatings were deposited from both ethanol and methanol solutions containing 3 weight percent of colloidal silica particles 10-20 nm in diameter[1]. A substrate was lowered vertically into the solution and then withdrawn at 4.0 cm/min. Build-up of coating thickness was accomplished by use of three depositions. The substrates were 2-inch diameter disks of Corning 7940 high purity fused silica that had been polished by Zygo, Inc. Immediately prior to application of the coating solution, the surfaces of these samples were cleaned with lens tissue and ethanol. Four of the substrates were also etched in dilute HF prior to being coated: additional comments on this etching are given below.

The two-surface transmittance spectrum for a silica substrate bearing three-layer sol-gel coatings is given in Fig. 1. The measured transmittance was .994 at 1064 nm, .996 at the spectral maximum near 1000 nm, and equal to that of the bare substrate at the minimum near 500 nm. Using the values of these transmittances and the positions of the spectral extrema, we estimate that the coatings were homogeneous layers

with optical thickness of 250 nm, refractive index ∿ 1.26, and silica content of 56%. The homogeneity of the coating suggests that dried porous material from an initial deposition was not filled during a subsequent deposition, and that the optical thickness was increased by about 83 nm during each application of the coating. A loss believed to be scattering caused transmittance at wavelengths below 250 nm to be less than expected.

3. Measurement of Laser Damage Thresholds

Damage tests were made with 1064-nm pulses in a weakly convergent beam that was about 2 mm in diameter at the sample surface. For each pulse used, the pulse energy and the energy distribution in the beam were measured, and the peak on-axis fluence was computed to within ± 7%.

Duration of the laser pulses was controlled by using a Pockels cell shutter to slice them from a Gaussian waveform pulse with duration of 150 ns. At the minimum pulse duration, 1 ns, the pulse waveform was governed by the rise and fall times for the shutter and was approximately Gaussian. Pulses with duration 5.5 and 9 ns had rise and fall times identical to those of the 1-ns pulses. These longer pulses were sliced from the leading edges of the 150-ns pulses and had waveforms that were trapezoidal with a slight upramp. Gain saturation in the laser amplifier reshaped the pulses into trapezoids with trailing edges as much as 30% below the leading edges, the saturation and reshaping being most significant in the pulses with greatest energy.

During test of a sample, each test site was irradiated only once. To detect damage, we observed for laser-induced light emission during a

shot, visually inspected the test site under intense white light illumination after the shot, and also used dark field microscopy to record 100 times magnified photographs of the center of the test site before and after the site was irradiated. Damage was defined to be a permanent alteration of the sample detectable by these examinations. The threshold-level damage usually consisted of very small volumes visible in the dark field presentation of the microscope. Except for the least damages, these areas were usually seen in the simple visual inspection of the test site, but were not always apparent in inspection by either white-light or Nomarski microscopy. Therefore we assume the damages were small localized fractures in the coating that were most apparent by their ability to scatter light.

Laser-induced emission of light was observed on some samples, particularly those with the greatest thresholds, during shots at fluences below those which caused permanent damage detectable by the inspections described above. At these subthreshold-fluences, the light emission did not occur as a bright spark. Instead, it appeared as a weakly luminous disk on the sample surface, had the size and shape of the laser beam, and sometimes abated when a site was repeatedly irradiated. It has not been determined whether this emission of light signals the existence of a plasma with density sufficient to cause a serious perturbation of a beam.

4. Experimental Results

Measured thresholds are given in Table 1. For coatings in the first two preparation groups, thresholds for those made from an ethanol solution were systematically above thresholds of those made from a

methanol solution, and thresholds of the methanol-solution coatings were almost independent of pulse duration. This was difficult to understand. Coatings made from the ethanol solution consist of layers of silica spheres having diameters of about 20 nm, whereas those made from the methanol solution consist of smaller spheres with diameters of 6-10 nm. Since the sizes of the spheres are all very small relative to the 1064 nm test wavelength, and since both of the precursor solutions should have provided silica of comparable purity, it was believed that the apparent difference in performance of the two types of coatings was due to an extrinsic effect.

Therefore, we fabricated a third set of coatings on the four substrates that had been used in fabrication of the initial group of coatings. The substrates were etched in dilute HF acid to remove the existing coatings, cleaned in water and ethanol, and recoated using the same ethanol and methanol solutions that had been used in preparation of coatings in sets 1 and 2.

Thresholds for the coatings in set 3 were independent of the type of precursor solution, systematically larger than those obscured in tests of coatings in sets 1 and 2, and they all scaled uniformly with pulse duration. However, for these latter coatings, thresholds for laser-induced light emission were sometimes significantly below the large fluences required to cause detectable permanent physical damage. The fluence for laser-induced emission of light may be the practical upper bound for thresholds in these coatings.

In Fig. 2 we have plotted all the measured thresholds as a function of pulse duration, indicated the median threshold at each pulse duration,

and also plotted the function $T = 10.8\tau^{.56}$ which is a best fit for scaling of the median threshold T with pulse duration τ. Note that the median thresholds are generally greater than thresholds for the initial methanol-sol films that may have been limited by substrate cleanliness, but well below the largest thresholds measured for coatings in set 3, and even conservative relative to light emission thresholds for coatings in set 3. Therefore, we believe it is possible by simple attention to preparation to reproducibly prepare coatings with thresholds greater than or equal to the median values.

The scaling of median thresholds as $\tau^{0.56}$ is within error comparable to the $\tau^{.5}$ scaling that has been observed in similar 1064-nm tests with bare polished glass surfaces[2] and with graded-index surfaces made by etching[3]. However, since the rule $\tau^{.5}$ can be attributed to a wide class of physical effects, it does not uniquely identify the mechanism for damage.

These coatings will be useful at a few places in the Nova amplifier chains. Lenses and windows that transmitted only the 1064-nm beams in Nova were made of BK-7 glass and had antireflective surfaces made by neutral solution processing (NSP)[4]. During initial operation of the Novette laser, transmittance of large diameter beams of high-intensity through optical elements made of BK-7 glass revealed the presence of a low volume density of platinum inclusions with damage thresholds of 4-7 J/cm^2 (1-ns, 1064-nm). Therefore, high purity fused silica may be more suitable than BK-7 glass for fabrication of a few elements in NOVA that receive the greatest fluence loading. In Fig. 3, we compare the 1-ns 1064-nm thresholds reported here for sol-gel AR films with thresholds

previously measured on NSP surfaces on Bk-7 glass. Thresholds for films made by the two process are comparable, so if we are required to use fused silica for some 1064-nm components in Nova, the sol-gel coatings should adequately serve as damage resistant AR films.

Acknowledgments

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References

1. Ian M. Thomas, John G. Wilder, W. Howard Lowdermilk, and M. C. Staggs, "High Damage Threshold Porous Silica Antireflection Coating," elsewhere in these proceedings.

2. David Milam, "1064 nm Laser-Damage Thresholds of Polished Glass Surfaces as a Function of Pulse Duration and Surface Roughness", in Laser Damage in Optical Materials: 1978, NBS Spec. Publ. 541, pp. 164-167, issued Dec. 1978.

3. James E. Swain, W. Howard Lowdermilk, and David Milam, "Raising the Surface Damage Threshold of Neutral Solution Processed BK-7 by Pulse Laser Irradiation," in Laser Induced Damage in Optical Materials: 1982, NBS Spec. Publ. 669, pp. 292-295, issued Jan. 1984.

4. Lee M. Cook, W. Howard Lowdermilk, David Milam, and James E. Swain, "Antireflective Surfaces for High-Energy Laser Optics formed by Neutral-Solution Processing," Appl. Optics, Vol. 21, p. 482-1485, April 15, 1982.

Table 1. 1064-nm Laser-Damage Thresholds of Sol-Gel AR Coatings on Fused Silica

Preparation Sample	Group	Coating Type	Laser-Damage Thresholds, J/cm^2					
			1 ns		5.5 ns		9 ns	
			damage	light	damage	light	damage	light
3811	1	Ethanol Sol	10 ± 1.5	< 8	27 ± 4	19 ± 3	36 ± 5	32 ± 11
3815	1	"	13 ± 2	5.8 ± 6	26 ± 4	21 ± 2	41 ± 6	34 ± 5
3810	1	Methanol Sol	9.4 ± 1.4	8.0 ± 2	16 ± 2.4	11 ± 1	17 ± 2.5	15 ± 3
3817	1	"	7.7 ± 2	13 ± 3	16 ± 4.5	> 31	11 ± 2	> 15
3746	2	"	3.4 ± 3	8 ± 2	7.6 ± 1	14 ± 1	6.7 ± 1	> 11
3744	2	Ethanol Sol	7 ± 1.2	7 ± 1.2	19 ± 2.8	18 ± 1.8	28 ± 4	16 ± 1.6
3815	3	Methanol Sol	18 ± 2.7	12 ± 1	46 ± 7	30 ± 1	61 ± 9	37 ± 15
3817	3	"	17 ± 2.6	12 ± 2	42 ± 6	33 ± 6	70 ± 10	50 ± 15
3810	3	Ethanol Sol	11 ± 1.7	11 ± 2	45 ± 7	43 ± 4	62 ± 10	53 ± 6
3811	3	"	13 ± 2	13 ± 1	48 ± 8	32 ± 3	62 ± 4	32 ± 3

Figure Captions

1. Two-surface transmittance for a silica window with sol-gel AR coatings on both surfaces. Dashed curve is the spectrum corrected for small measurement errors.

2. 1064-nm damage thresholds of sol-gel AR coatings as a function of laser pulse duration. Dashed line is the curve $T = 10.8\ \tau^{0.56}$ which describes scaling of the median thresholds.

3. Laser-damage thresholds measured with 1-ns, 1064-nm pulses on sol-gel AR coatings (top) and on surfaces made by neutral solution processing (bottom).

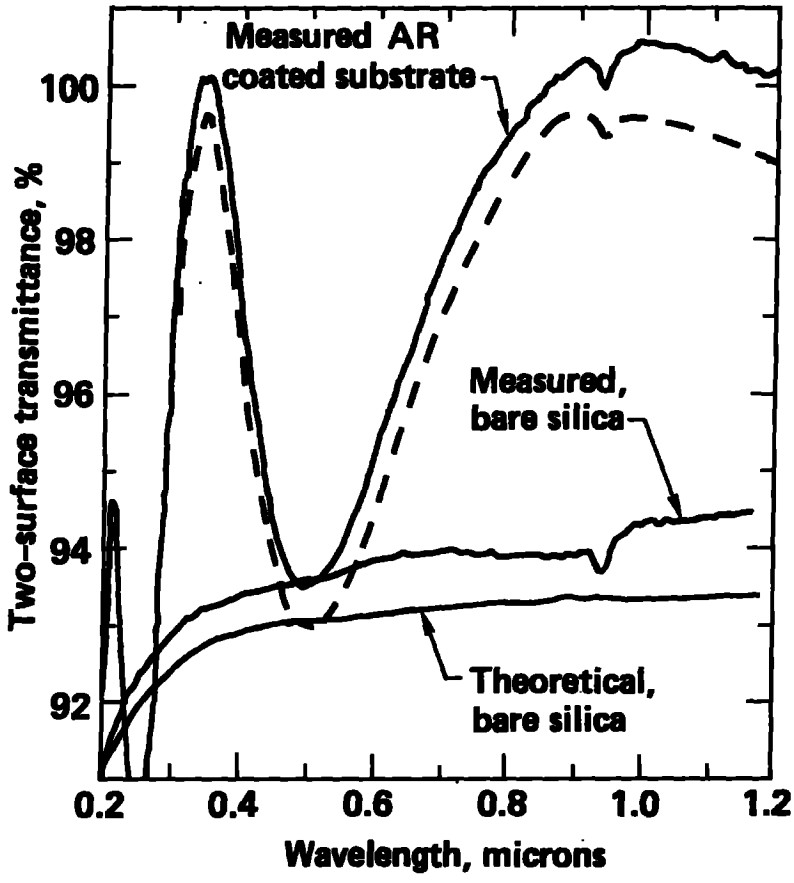

Measured AR
coated substrate
Measured,
bare silica
Theoretical,
bare silica
Two-surface transmittance, %
100
98
96
94
92
0.2
0.4
0.6
0.8
1.0
1.2
Wavelength, microns

Figure 1

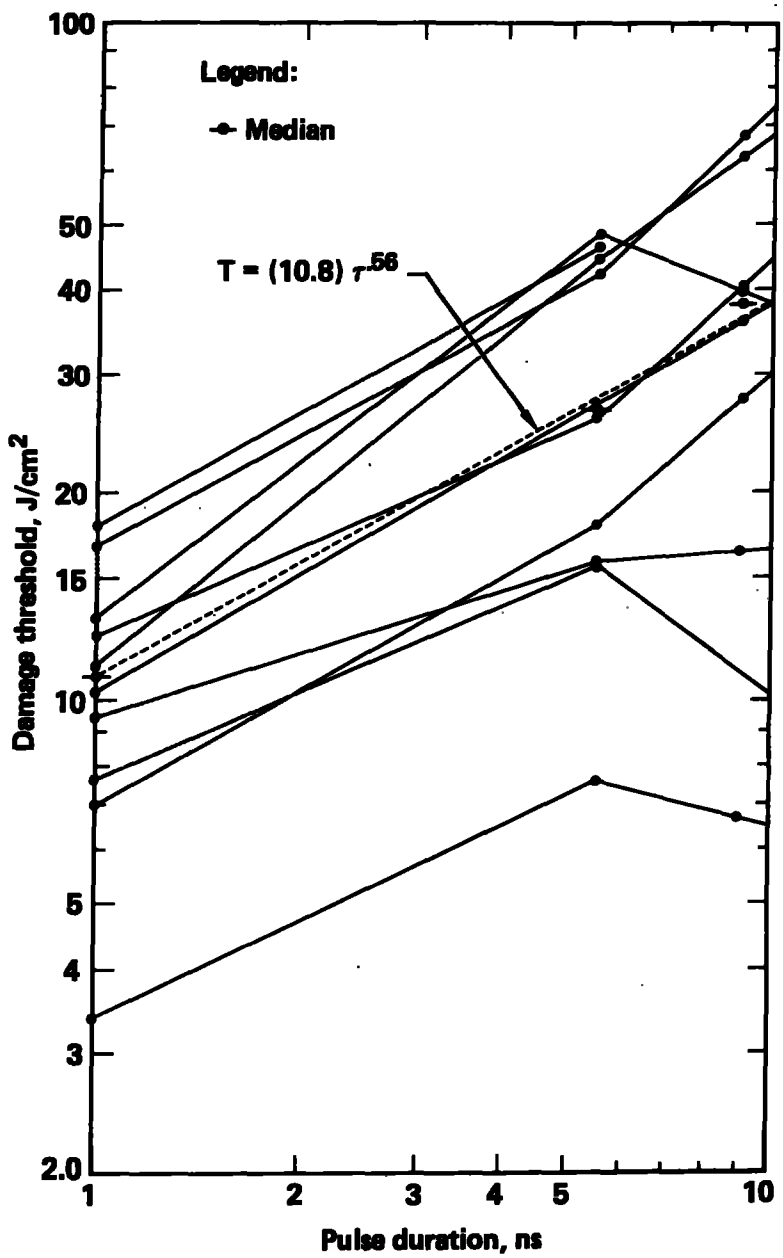
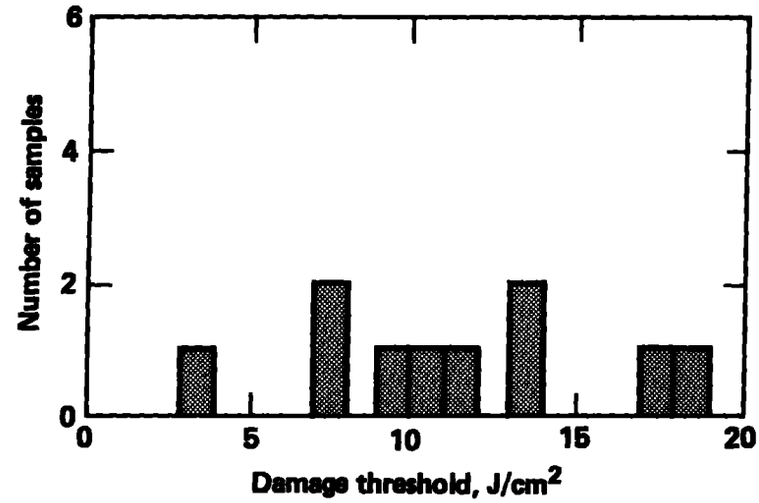

Legend:
Median
T = (10.8) τ^.56
Damage threshold, J/cm²
Pulse duration, ns
100
50
40
30
20
15
10
5
4
3
2.0
1
2
3
4
5
10

Figure 2


Number of samples
0
2
4
6
0
5
10
15
20
Damage threshold, J/cm2

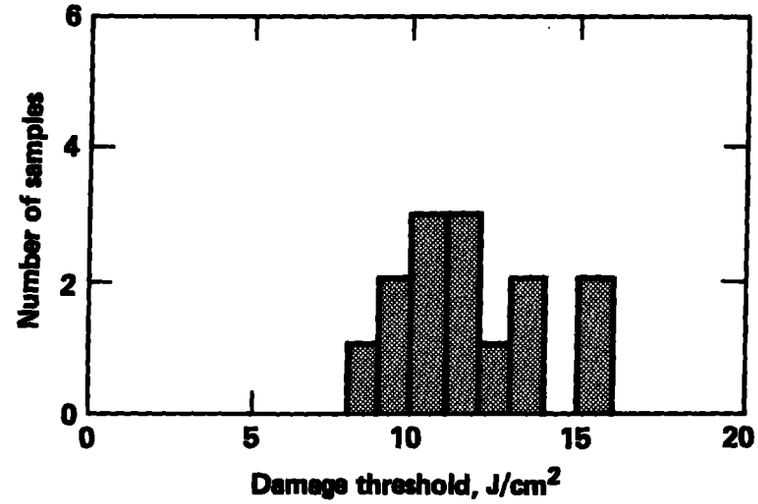

Number of samples
0
2
4
6
0
5
10
15
20
Damage threshold, J/cm2

Figure 3